



Europäisches  
Patentamt

European  
Patent Office

Office européen  
des brevets

REC'D 23 OCT 2003

WIPO

PCT

Bescheinigung

Certificate

Attestation

Die angehefteten Unterla-  
gen stimmen mit der  
ursprünglich eingereichten  
Fassung der auf dem näch-  
sten Blatt bezeichneten  
europäischen Patentanmel-  
dung überein.

The attached documents  
are exact copies of the  
European patent application  
described on the following  
page, as originally filed.

Les documents fixés à  
cette attestation sont  
conformes à la version  
initialement déposée de  
la demande de brevet  
européen spécifiée à la  
page suivante.

Patentanmeldung Nr. Patent application No. Demande de brevet n°

02021421.9

**PRIORITY  
DOCUMENT**  
SUBMITTED OR TRANSMITTED IN  
COMPLIANCE WITH RULE 17.1(a) OR (b)

Der Präsident des Europäischen Patentamts;  
Im Auftrag

For the President of the European Patent Office

Le Président de l'Office européen des brevets  
p.o.

R C van Dijk

BEST AVAILABLE COPY



Anmeldung Nr:  
Application no.: 02021421.9  
Demande no:

Anmeldetag:  
Date of filing: 25.09.02  
Date de dépôt:

Anmelder/Applicant(s)/Demandeur(s):

Basell Poliolefine Italia S.p.A.  
Via Pergolesi 25  
20124 Milano  
ITALIE

Bezeichnung der Erfindung/Title of the invention/Titre de l'invention:  
(Falls die Bezeichnung der Erfindung nicht angegeben ist, siehe Beschreibung.  
If no title is shown please refer to the description.  
Si aucun titre n'est indiqué se référer à la description.)

Polypropylene fibres suitable for spunbonded non-woven fabrics

In Anspruch genommene Priorität(en) / Priority(ies) claimed /Priorité(s)  
revendiquée(s)

Staat/Tag/Aktenzeichen/State/Date/File no./Pays/Date/Numéro de dépôt:

Internationale Patentklassifikation/International Patent Classification/  
Classification internationale des brevets:

D01F6/46

Am Anmeldetag benannte Vertragstaaten/Contracting states designated at date of  
filing/Etats contractants désignées lors du dépôt:

AT BE BG CH CY CZ DE DK EE ES FI FR GB GR IE IT LI LU MC NL PT SE SK TR

25. Sep. 2002

**"POLYPROPYLENE FIBRES SUITABLE FOR SPUNBONDED NON-WOVEN FABRICS"**

The present invention relates to thermal bondable fibres comprising propylene polymer compositions, spunbonded non-woven fabrics obtained from the said fibres and polypropylene compositions for the production of the said fibres.

The definition for fibres includes spunbonding fibres.

Fibres formed with olefin copolymers or polyolefin blends are already known in the art. In particular, the use of random copolymers of propylene has already been contemplated just to improve the thermal bondability (i.e. the bond strength) of fibres and/or the calendering speed.

Propylene random copolymer fibres also are typically used for non-woven fabrics to improve the non-woven softness, and good properties are generally obtained at elevated contents of the soluble fraction. However, the drawback is that elevated solvent-soluble contents substantially reduce the non-woven tenacity of the non-woven fabric.

Example of the fibres formed with olefin copolymers and polyolefin blends are described in European patent 416 620. The said polymers that have a crystallinity less than 45% provide fibres have lower tenacity and lower modulus than those formed from more crystalline propylene polymers but with improved softness and maintaining fabric characteristics.

Another example is U.S. patent 4,211,819, describing heat-melt adhesive fibres obtained by spinning a crystalline (propylene-co-ethylene-co-1-butene)polymer. Such fibres having a lower heat-melting adhesion temperature are used as binder material only, therefore the mechanical properties are conferred by other materials. In fact, when non-woven fabrics are prepared in the examples, the said fibres are mixed with rayon fibres before calendering.

The applicant has now found that thanks to particular olefin copolymers and olefin copolymer blends, fibres having an improved thermal bondability associated with a good balance of mechanical properties, and non-woven fabrics having a better balance of thermal bonding characteristics and physical properties can be provided. Thus, at the same or even lower thermal bonding temperature, the non-woven fabric of the present invention exhibits improved properties such as tenacity and elongation in comparison with known spunbonded non-woven fabrics made of propylene homopolymers and copolymers with similar melt flow rate (MFR).

Another advantage of the spunbonded non-woven fabrics is the improved softness. The

higher softness contributes, with the soft touch, to improve the final quality of the non-woven fabric, in particular for the hygiene applications where the market appreciates very soft non-woven fabrics with clothlike appearance.

A further advantage is that by properly selecting some intrinsic properties of the propylene polymer material, non-woven fabrics with excellent properties, such as tenacity even in combination with high elongation, and low solvent-soluble contents are obtained.

Therefore the present invention provides fibres for spun bonding comprising a propylene polymer composition (A) having an MFR L (1) value from 6 to 60 g/10 min, preferably from 10 to 40 g/10 min, selected from the group consisting of:

- i) a crystalline propylene random copolymer containing at least 0.8% by weight of ethylene and optionally one or more C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefins and having a melting temperature of 155° C or higher, a content of fraction soluble in xylene at room temperature (about 25° C) lower than 5% by weight, a value of the ratio of the polymer fraction collected at the temperature range from 25° to 95° C by temperature rising elution fractionation with xylene (TREF) to the xylene soluble fraction (at 25° C), higher than 8; and
- ii) a crystalline propylene polymer blend containing at least 0.64 wt% of ethylene recurring unit and comprising (percent by weight):
  - I) 20-80%, preferably 30-70%, of a crystalline propylene homopolymer or crystalline propylene random copolymer containing up to 1.5% by weight of ethylene; and
  - II) 20-80%, preferably 30-70%, of a crystalline propylene random copolymer selected from the group consisting of:
    - IIa) a crystalline copolymer of propylene with 0.8 to 9% by weight of ethylene; provided that the difference in the ethylene content between polymer (I) and polymer (IIa) be at least 0.8 percentage unit, preferably 1 percentage unit, more preferably 2 percentage units, with respect to the weight of the (co)polymer concerned;
    - IIb) a crystalline copolymer of propylene with 1.5 to 18% by weight of a C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefin and optionally ethylene; provided that the difference in the comonomer content between polymer (I) and polymer (IIb) be at least 1.5 percentage units, preferably 2 percentage units, with respect to the weight of the (co)polymer concerned; and

IIc) a mixture of copolymer (IIa) and copolymer (IIb).

Preferably, the blend (ii) of said polymer composition (A) has the following features:

- 1) a melting temperature of 153° C or higher; and
- 2) a content of fraction soluble in xylene at room temperature (about 25° C) lower than 8% by weight, preferably lower than 5% by weight, more preferably lower than 2.5% by weight.

The above crystalline polymers have a stereoregularity of the isotactic type.

When only ethylene is present as the comonomer in copolymer (i), it is generally within 0.8 and 2% by weight with respect to the weight of the polymer.

When C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefins also are present, they are generally within 1 and 6% by weight with respect to the weight of the polymer.

When ethylene also is present as the comonomer in random copolymer (IIb), the ethylene content is generally within 1% by weight with respect to the weight of the polymer.

In a preferred embodiment of composition (A) the polymer blend (ii) comprises (percent by weight):

- I) 20-80%, preferably 30-70%, of a crystalline propylene homopolymer and/or crystalline propylene random copolymer containing up to 1.5% by weight of ethylene; and
- II) 20-80%, preferably 30-70%, of a crystalline random copolymer selected from:
  - IIa) a copolymer of propylene with 0.8 to 5% by weight of ethylene; provided that the difference in the ethylene content between polymer (I) and polymer (IIa) be at least 0.8 percentage unit with respect to the weight of the (co)polymer concerned;
  - IIb) a copolymer of propylene with 1.5 to 12% by weight of a C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefin and optionally ethylene; provided that the difference in the comonomer content between polymer (I) and polymer (IIb) be at least 1.5 percentage units with respect to the weight of the (co)polymer concerned; and
  - IIc) a mixture of copolymer (IIa) and copolymer (IIb).

The said blend (ii) has preferably a melting temperature of 155° C or higher, a content of fraction soluble in xylene at room temperature lower than 5% by weight and a value of the ratio of the polymer fraction collected at the temperature range from 25° to 95° C by temperature rising elution fractionation with xylene to the xylene soluble fraction higher (at 25° C) than 8.

Particularly preferred is a crystalline propylene polymer blend (ii) wherein component (I) is a propylene homopolymer and component (II) is an ethylene-propylene random copolymer. Spun-bonded non-woven fabrics having particularly high values of both tenacity and elongation at break are typically obtained from fibres prepared with compositions (A) having an MFR L (1) value from 10 to 40 g/10 min, preferably 20 to 40 g/10 min. Preferably, said composition (A) has a polydispersity index (PI) value is typically from 3 to 6, more preferably 3.5 to 6. The spun-bonded non-woven fabric produced with fibres comprising said compositions (A) have typically a tenacity value of 90 N/5cm measured in machine direction and of 60 N/5cm measured in cross direction. The elongation at break is typically at least 90%, preferably 100%, measured in both the directions.

Also suitable fibres for spun bonding are prepared with said polypropylene composition (A) obtainable by subjecting to chemical degradation a precursor polymer composition (B) having an MFR L value (MFR L (2)) of from 0.5 to 6 g/10 min preferably 0.5 to 3.5 g/10 min, provided that the ratio MFR L (1) to MFR L (2) be from 1.5 to 60, preferably 6 to 30. Preferably, said chemical degraded composition (A) has a polydispersity index (PI) value from 2 to 6, more preferably from 2 to 3.

The fibre according to the present invention has typically a value of tenacity higher than 22 cN/tex, preferably higher than 23 cN/tex, with standard throughput.

The elongation at break shown by the fibres of the present invention is typically higher than 140%, preferably at least 150%, with standard throughput

The fibre according to the present invention has typically a titre ranging from 0.8 and 8 dtex. The chemical degradation of the polymer chains of the precursor polymer (B) is carried out by using appropriate and known techniques.

One of said techniques is based on the use of peroxides which are added in an extruder to the polymeric material in a quantity that allows to obtain the desired degree of chemical degradation. Such degradation is achieved by bringing the polymeric material at a temperature at least equal to the decomposition temperature of the peroxides and mechanical shear stress conditions.

The peroxides that are most conveniently employable for the chemical degradation have a decomposition temperature preferably ranging from 150 to 250°C. Examples of said peroxides are the di-tert-butyl peroxide, the dicumyl peroxide, the 2,5-dimethyl-2,5-di (tert-butyl peroxy) hexyne, and the 2,5-dimethyl-2,5-di (tert-butyl peroxy) hexane, which is

marketed under the Luperox 101 trade name.

The MFR values of the two propylene polymers constituting polymer blend (ii) can be similar or different. In a particular embodiment of the present invention the MFR value of propylene polymers (Ia) and (IIa) is lower than that of propylene random copolymers (Ib) and (IIb), respectively.

The C<sub>4</sub>-C<sub>10</sub> α-olefins, that may be present as comonomers in the said propylene polymer or polymer composition, are represented by the formula CH<sub>2</sub>=CHR, wherein R is an alkyl radical, linear or branched, with 2-8 carbon atoms or an aryl (in particular phenyl) radical. Examples of said C<sub>4</sub>-C<sub>10</sub> α-olefins are 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene and 1-octene. Particularly preferred is 1-butene.

Therefore, a further embodiment of the present invention is a chemically degraded propylene polymer composition (A) having an MFR L (1) value from 6 to 60 g/10 min, preferably from 10 to 40 g/10 min, and being selected from:

- i) a crystalline propylene random copolymer containing at least 0.8% by weight of ethylene and optionally one or more C<sub>4</sub>-C<sub>10</sub> α-olefins; and
- ii) a crystalline propylene polymer blend consisting of (percent by weight):
  - I) 20-80%, preferably from 30-70%, of a crystalline propylene homopolymer or crystalline propylene random copolymer containing up to 1.5% by weight of ethylene; and
  - II) 20-80%, preferably from 30-70%, of a crystalline propylene random copolymer selected from:
    - IIa) a copolymer of propylene with 0.8 to 5% by weight of ethylene; provided that the difference in the ethylene content between polymer (I) and polymer (IIa) be at least 0.8 percentage unit, preferably 1 percentage unit, more preferably 2 percentage units, with respect to the weight of the (co)polymer concerned;
    - IIb) a copolymer of propylene with 1.5 to 12% by weight of a C<sub>4</sub>-C<sub>10</sub> α-olefin and optionally ethylene; provided that the difference in the comonomer content between polymer (I) and polymer (IIb) be at least 1.5 percentage units, preferably 2 percentage units, with respect to the weight of the (co)polymer concerned; and
    - IIc) a mixture of copolymer (IIa) and copolymer (IIb).

The said polymer composition is obtainable by subjecting to chemical degradation a precursor polymer composition (B) having an MFR L value (MFR L (2)) of from 0.5 to 6 g/10 min, preferably from 0.5 to 3.5 g/10 min, provided that the ratio MFR L (1) to MFR L (2) be from 1.5 to 60, preferably from 6 to 30.

The said polymer composition (A) has preferably the following features:

- 1) a melting temperature of 155° C or higher;
- 2) a content of fraction soluble in xylene at room temperature (about 25° C) lower than 4% by weight, preferably lower than 3% by weight, more preferably lower than 2.5% by weight; and
- 3) a value of the ratio of the polymer fraction collected at the temperature range from 25° to 95° C by temperature rising elution fractionation with xylene (TREF) to the xylene soluble fraction, higher than 8 wt%, preferably higher than 10 wt%, more preferably higher than 12 wt%.

Preferably, said composition (A) has a PI value from 2.0 to 4.5, more preferably from 2.0 to 3.0.

The propylene polymer composition (A) can be prepared with a process comprising the following stages:

- 1) preparing the previously said precursor composition (B) by polymerising the monomers in one or more sequential steps, operating in each step in the presence of the polymer formed and the catalyst used in the preceding step, and dosing the molecular weight regulator (preferably hydrogen) in such amounts as to obtain an MFR L (2) value for the precursor composition of from 0.5 to 6 g/10 min, preferably from 0.5 to 3.5 g/10 min; and
- 2) subjecting the precursor composition (B) obtained in 1) to a degradation treatment in order to obtain the said MFR L (1) values for the final composition from 6 to 60 g/10 min, more preferably from 10 to 40 g/10 min, with a degradation ratio, in terms of ratio MFR L (1) to MFR L (2), of from 1.5 to 60, preferably from 6 to 30.

Such a preferred process is extremely convenient, as it avoids the separate preparation of the precursor composition and degradation treatments.

From the description above it should be clear that in the precursor composition (B) the comonomer contents and relative amounts are the same as in the final composition (A) (after degradation). The degradation treatment has the effect of increasing the MFR L values of the



composition from MFR L (2) to MFR L (1), with the said values of the ratio between the two MFR L values, namely MFR L (1)/MFR L (2), from 1.5 to 60, preferably from 6 to 30.

The compositions of the present invention can be prepared by polymerisation in one or more polymerisation steps. Such polymerisation is carried out in the presence of stereospecific Ziegler-Natta catalysts. An essential component of said catalysts is a solid catalyst component comprising a titanium compound having at least one titanium-halogen bond, and an electron-donor compound, both supported on a magnesium halide in active form. Another essential component (co-catalyst) is an organoaluminum compound, such as an aluminum alkyl compound.

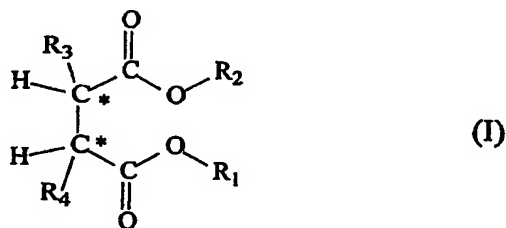
An external donor is optionally added.

The catalysts generally used in the process of the invention are capable of producing polypropylene with an isotacticity index greater than 90%, preferably greater than 95%.

Catalysts having the above mentioned characteristics are well known in the patent literature; particularly advantageous are the catalysts described in US patent 4,399,054 and European patent 45977. Other examples can be found in US patent 4,472,524.

The solid catalyst components used in said catalysts comprise, as electron-donors (internal donors), compounds selected from the group consisting of ethers, ketones, lactones, succinates, compounds containing N, P and/or S atoms, and esters of mono- and dicarboxylic acids.

Particularly suitable electron-donor compounds are non-extractable succinates; particularly preferred are the succinates of formula (I) below



in which the radicals  $R_1$  and  $R_2$ , equal to, or different from, each other are a  $C_1$ - $C_{20}$  linear or branched alkyl, alkenyl, cycloalkyl, aryl, arylalkyl or alkylaryl group, optionally containing heteroatoms; and the radicals  $R_3$  and  $R_4$  equal to, or different from, each other, are  $C_1$ - $C_{20}$  alkyl, cycloalkyl, aryl, arylalkyl or alkylaryl group, optionally containing heteroatoms with the proviso that at least one of them is a branched alkyl; said compounds being, with respect

to the two asymmetric carbon atoms identified in the structure of formula (I), stereoisomers of the type (S,R) or (R,S) that are present in pure forms or in mixtures.

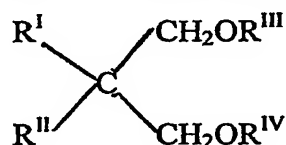
R<sub>1</sub> and R<sub>2</sub> are preferably C<sub>1</sub>-C<sub>8</sub> alkyl, cycloalkyl, aryl, arylalkyl and alkylaryl groups.

Particularly preferred are the compounds in which R<sub>1</sub> and R<sub>2</sub> are selected from primary alkyls and in particular branched primary alkyls. Examples of suitable R<sub>1</sub> and R<sub>2</sub> groups are methyl, ethyl, n-propyl, n-butyl, isobutyl, neopentyl, 2-ethylhexyl. Particularly preferred are ethyl, isobutyl, and neopentyl.

Particularly preferred are the compounds in which the R<sub>3</sub> and/or R<sub>4</sub> radicals are secondary alkyls like isopropyl, sec-butyl, 2-pentyl, 3-pentyl or cycloalkyls like cyclohexyl, cyclopentyl, cyclohexylmethyl.

Examples of the above-mentioned compounds are the (S,R) (S,R) forms pure or in mixture, optionally in racemic form, of diethyl 2,3-bis(trimethylsilyl)succinate, diethyl 2,3-bis(2-ethylbutyl)succinate, diethyl 2,3-dibenzylsuccinate, diethyl 2,3-diisopropylsuccinate, diisobutyl 2,3-diisopropylsuccinate, diethyl 2,3-bis(cyclohexylmethyl)succinate, diethyl 2,3-diisobutylsuccinate, diethyl 2,3-dineopentylsuccinate, diethyl 2,3-dicyclopentylsuccinate, diethyl 2,3-dicyclohexylsuccinate.

Other suitable electron-donor compounds are 1,3-diethers of formula:



wherein R<sup>I</sup> and R<sup>II</sup> are the same or different and are C<sub>1</sub>-C<sub>18</sub> alkyl, C<sub>3</sub>-C<sub>18</sub> cycloalkyl or C<sub>7</sub>-C<sub>18</sub> aryl radicals; R<sup>III</sup> and R<sup>IV</sup> are the same or different and are C<sub>1</sub>-C<sub>4</sub> alkyl radicals; or are the 1,3-diethers in which the carbon atom in position 2 belongs to a cyclic or polycyclic structure made up of 5, 6, or 7 carbon atoms, or of 5-n or 6-n' carbon atoms, and respectively n nitrogen atoms and n' heteroatoms selected from the group consisting of N, O, S and Si, where n is 1 or 2 and n' is 1, 2, or 3, said structure containing two or three unsaturations (cyclopolyenic structure), and optionally being condensed with other cyclic structures, or substituted with one or more substituents selected from the group consisting of linear or branched alkyl radicals; cycloalkyl, aryl, aralkyl, alkaryl radicals and halogens, or being condensed with other cyclic structures and substituted with one or more of the above mentioned substituents that can also be bonded to the condensed cyclic structures; one or more of the above mentioned alkyl, cycloalkyl, aryl, aralkyl, or alkaryl radicals and the

condensed cyclic structures optionally containing one or more heteroatoms as substitutes for carbon or hydrogen atoms, or both.

Ethers of this type are described in published European patent applications 361493 and 728769.

Representative examples of said dieters are 2-methyl-2-isopropyl-1,3-dimethoxypropane, 2,2-diisobutyl-1,3-dimethoxypropane, 2-isopropyl-2-cyclopentyl-1,3-dimethoxypropane, 2-isopropyl-2-isoamyl-1,3-dimethoxypropane, 9,9-bis (methoxymethyl) fluorene.

Other suitable electron-donor compounds are phthalic acid esters, such as diisobutyl, dioctyl, diphenyl and benzylbutyl phthalate.

The preparation of the above mentioned catalyst components is carried out according to various methods.

For example, a  $\text{MgCl}_2 \cdot n\text{ROH}$  adduct (in particular in the form of spheroidal particles) wherein  $n$  is generally from 1 to 3 and ROH is ethanol, butanol or isobutanol, is reacted with an excess of  $\text{TiCl}_4$  containing the electron-donor compound. The reaction temperature is generally from  $80^\circ$  to  $120^\circ$  C. The solid is then isolated and reacted once more with  $\text{TiCl}_4$ , in the presence or absence of the electron-donor compound, after which it is separated and washed with aliquots of a hydrocarbon until all chlorine ions have disappeared.

In the solid catalyst component the titanium compound, expressed as Ti, is generally present in an amount from 0.5 to 10% by weight. The quantity of electron-donor compound which remains fixed on the solid catalyst component generally is 5 to 20% by moles with respect to the magnesium dihalide.

The titanium compounds which can be used for the preparation of the solid catalyst component are the halides and the halogen alcoholates of titanium. Titanium tetrachloride is the preferred compound.

The reactions described above result in the formation of a magnesium halide in active form. Other reactions are known in the literature, which cause the formation of magnesium halide in active form starting from magnesium compounds other than halides, such as magnesium carboxylates.

The active form of magnesium halide in the solid catalyst component can be recognized by the fact that in the X-ray spectrum of the catalyst component the maximum intensity reflection appearing in the spectrum of the nonactivated magnesium halide (having a surface area smaller than  $3 \text{ m}^2/\text{g}$ ) is no longer present, but in its place there is a halo with the

maximum intensity shifted with respect to the position of the maximum intensity reflection of the nonactivated magnesium dihalide, or by the fact that the maximum intensity reflection shows a width at half-peak at least 30% greater than the one of the maximum intensity reflection which appears in the spectrum of the nonactivated magnesium halide. The most active forms are those where the above mentioned halo appears in the X-ray spectrum of the solid catalyst component.

Among magnesium halides, the magnesium chloride is preferred. In the case of the most active forms of magnesium chloride, the X-ray spectrum of the solid catalyst component shows a halo instead of the reflection which in the spectrum of the nonactivated chloride appears at 2.56 Å.

The Al-alkyl compounds used as co-catalysts comprise the Al-trialkyls, such as Al-triethyl, Al-triisobutyl, Al-tri-n-butyl, and linear or cyclic Al-alkyl compounds containing two or more Al atoms bonded to each other by way of O or N atoms, or SO<sub>4</sub> or SO<sub>3</sub> groups.

The Al-alkyl compound is generally used in such a quantity that the Al/Ti ratio be from 1 to 1000.

The electron-donor compounds that can be used as external donors include aromatic acid esters such as alkyl benzoates, and in particular silicon compounds containing at least one Si-OR bond, where R is a hydrocarbon radical.

Examples of silicon compounds are (tert-butyl)<sub>2</sub>Si(OCH<sub>3</sub>)<sub>2</sub>, (cyclohexyl)(methyl)Si(OCH<sub>3</sub>)<sub>2</sub>, (phenyl)<sub>2</sub>Si(OCH<sub>3</sub>)<sub>2</sub> and (cyclopentyl)<sub>2</sub>Si(OCH<sub>3</sub>)<sub>2</sub>. 1,3-diethers having the formulae described above can also be used advantageously. If the internal donor is one of these diethers, the external donors can be omitted.

In particular, even if many other combinations of the previously said catalyst components may allow to obtain polymers and polymer compositions having the previously said features 1) and 2), the random copolymers are preferably prepared by using catalysts containing a phthalate as inside donor and (cyclopentyl)<sub>2</sub>Si(OCH<sub>3</sub>)<sub>2</sub> as outside donor, or the said 1,3-diethers as inside donors and (cyclopentyl)<sub>2</sub>Si(OCH<sub>3</sub>)<sub>2</sub> as outside donor.

As previously said, the polymerisation process can be carried out in one or more steps. In the case of composition (II), it can be carried out in at least two sequential steps, wherein the first propylene (co)polymer and the second propylene random copolymer are prepared in separate subsequent steps, operating in each step, except the first step, in the presence of the polymer formed and the catalyst used in the preceding step. Clearly, when the composition

(II) contains additional (co)polymers, it becomes necessary to add further polymerisation steps to produce them. The said polymerisation steps can be carried out in separate reactors, or in one or more reactors where gradients of monomer concentrations and polymerisation conditions are generated. The catalyst is generally added only in the first step, however its activity is such that it is still active for all the subsequent step(s).

The regulation of the molecular weight is carried out by using known regulators, hydrogen in particular.

By properly dosing the concentration of the molecular weight regulator in the relevant steps, the previously described MFR values (MFR L (2)) are obtained.

The whole polymerisation process, which can be continuous or batch, is carried out following known techniques and operating in liquid phase, in the presence or not of inert diluent, or in gas phase, or by mixed liquid-gas techniques.

Reaction time, pressure and temperature relative to the two steps are not critical, however it is best if the temperature is from 20° to 100° C. The pressure can be atmospheric or higher.

The catalysts can be pre-contacted with small amounts of olefins (prepolymerisation).

It is also possible to employ a process for the catalytic polymerisation in the gas-phase carried out in at least two interconnected polymerisation zones, the process comprising feeding one or more monomers to said polymerisation zones in the presence of catalyst under reaction conditions and collecting the polymer product from said polymerisation zones, in which process the growing polymer particles flow upward through one of said polymerisation zones (riser) under fast fluidisation conditions, leave said riser and enter another polymerisation zone (downcomer) through which they flow downward under the action of gravity, leave said downcomer and are reintroduced into the riser, thus establishing a circulation of polymer between the riser and the downcomer, the process being optionally characterised in that:

- means are provided which are capable of totally or partially preventing the gas mixture present in the riser from entering the downcomer, and
- a gas and/or liquid mixture having a composition different from the gas mixture present in the riser is introduced into the downcomer.

Such polymerisation process is illustrated in WO 00/02929.

According to a particularly advantageous embodiment of this process, the introduction into the downcomer of the said gas and/or liquid mixture having a composition different from the

gas mixture present in the riser is effective in preventing the latter mixture from entering the downcomer.

The propylene polymer composition (A) used for fibres and non-woven fabrics of the present invention can also contain additives commonly employed in the art, such as antioxidants, light stabilizers, heat stabilizers, antistatic agents, flame retardants, fillers, nucleating agents, pigments, anti-soiling agents, photosensitizers.

As said above an embodiment of the present invention is the non-woven fabric. It has typically a weight below  $200 \text{ g/m}^2$ .

The fabrics of the present invention can be prepared with the well known processes for the preparation of spunbond non-woven fabrics, with which the fibres are spread to form directly a fibre web and calendered to obtain a non-woven fabric.

In a typical spunbonding process, the polymer is heated in an extruder to the melting point of the polymer or polymer blend and then the molten polymer is pumped under pressure through a spinneret containing a number of orifices of desired diameter, thereby producing filaments of the molten polymer and without subjecting the filaments to a subsequent drawing.

The equipment is characterised by the fact that it includes an extruder with a die on its spinning head, a cooling tower an air suction gathering device that uses Venturi tubes. Underneath this device, that uses air speed to control the take up speed, the filaments are usually gathered over a conveyor belt, where they are distributed forming a web for thermal bonding in a calender.

When using typical spunbonding machinery, it is usually convenient to apply the following process conditions:

- The output per hole ranges from 0.1 to 2 g/min, preferably from 0.2 to 1 g/min;
- The molten polymer filaments fed from the face of the spinneret are generally cooled by means of an air flow and are solidified as a result of cooling;
- The spinning temperature is generally between  $200^\circ$  and  $300^\circ \text{ C}$ , preferably between  $220^\circ$  and  $250^\circ \text{ C}$ .

The fabric can be constituted by monolayer or multilayer non-wovens.

In a preferred embodiment, at least one layer is substantially made of the said propylene polymer or propylene polymer composition.

Typically, the spun-bonded non-woven fabric made of fibres comprising chemical degraded composition (A) according to the present invention has a tenacity of at least 30 N measured in the machine direction and of at least 7.7 N measured in cross direction and an elongation at break of at least 38% measured in machine direction and of at least 58% measured in cross direction.

The non-woven fabric of the present invention can be multilayered and at least one layer comprises fibres formed from said propylene polymer composition (A). The other layer may be obtained by spinning processes other than spunbonding and may comprise other types of polymers.

The non-woven fabric of the present invention with improved tenacity, softness and elongation at break are useful in a number of applications. For example, the non-woven fabric can be converted into coverstock and diapers.

The following examples are given to illustrate and not to limit the present invention.

The data relating to the polymeric materials and the fibres of the examples are determined by way of the methods reported below.

- MFR: ISO 1133 (230° C, 2.16 kg);
- Melting and crystallization temperature: by DSC with a temperature variation of 20° C per minute;
- Ethylene content: by IR spectroscopy;
- Polydispersity Index (PI): measurement of molecular weight distribution of the polymer. To determine the PI value, the modulus separation at low modulus value, e.g. 500 Pa, is determined at a temperature of 200° C by using a RMS-800 parallel plates rheometer model marketed by Rheometrics (USA), operating at an oscillation frequency which increases from 0.01 rad/second to 100 rad/second. From the modulus separation value, the PI can be derived using the following equation:  

$$PI = 54.6 \times (\text{modulus separation})^{-1.76}$$
 wherein the modulus separation (MS) is defined as:  

$$MS = (\text{frequency at } G' = 500 \text{ Pa}) / (\text{frequency at } G'' = 500 \text{ Pa})$$
 wherein  $G'$  is the storage modulus and  $G''$  is the low modulus.
- Fractions soluble and insoluble in xylene at 25° C: 2.5 g of polymer are dissolved in 250 ml of xylene at 135° C under agitation. After 20 minutes the solution is allowed to cool to 25° C, still under agitation, and then allowed to settle for 30 minutes. The

precipitate is filtered with filter paper, the solution evaporated in nitrogen flow, and the residue dried under vacuum at 80° C until constant weight is reached. Thus one calculates the percent by weight of polymer soluble and insoluble at room temperature (25° C).

- Temperature rising elution fractionation with xylene (TREF): About 1 g of sample is dissolved in 200 mL of o-xylene, stabilized with 0.1 g/L of Iragnox® 1010 (pentaerythrityl tetrakis 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propanoate). The dissolution temperature is in the range of 125-135° C. The resulting solution is poured off into a column packed with glass beads and subsequently cooled down slowly in 16.5 h to 25° C.

The first fraction is obtained at room temperature eluting with o-xylene. The second fraction is collected after having raised the column temperature up to 95° C. The polymer component soluble between 25° and 95° C is collected as a single fraction.

The successive fractions are eluted with o-xylene while the temperature is raised linearly between 95° to 125° C. Each fraction, recovered as a 200 mL solution, is collected at 1° C temperature increments. The polymer fractions are subsequently precipitated with acetone, filtered on a 0.5 µm PTFE filter, dried under vacuum at 70° C and weighted.

- Titre of fibres: from a 10 cm long roving, 50 fibres are randomly chosen and weighed. The total weight of the said 50 fibres, expressed in mg, is multiplied by 2, thereby obtaining the titre in dtex.

- Tenacity and Elongation (at break) of fibres: from a 500 m roving a 100 mm long segment is cut. From this segment the single fibres to be tested are randomly chosen. Each single fibre to be tested is fixed to the clamps of an Instron dynamometer (model 1122) and tensioned to break with a traction speed of 20 mm/min for elongations lower than 100% and 50 mm/min for elongations greater than 100%, the initial distance between the clamps being of 20 mm. The Ultimate strength (load at break) and the Elongation at break are determined.

The Tenacity is derived using the following equation:

$$\text{Tenacity} = \text{Ultimate strength (cN)} \times 10 / \text{Titre (dtex)}$$

- Bond strength: when non-woven samples are prepared, the bond strength is determined on specimens 20 cm long and 5 cm wide. The 5 cm wide extremities are fixed to the



clamps of the dynamometer and tensioned at a clamp speed of 100 mm/min (the initial distance between the clamps being of 10 cm). The maximum force measured in the machine direction (MD) and in the cross direction (CD), with respect to the calendering step, represents the strength of the fibres.

#### Examples 1 and 2

The polymers are prepared by polymerising propylene and ethylene under continuous conditions in a plant comprising a gas phase polymerisation apparatus.

The catalyst employed comprises a catalyst component prepared by analogy with Example 5 of EP A 728 769, but using microspheroidal  $\text{MgCl}_2 \cdot 1.7 \text{ C}_2\text{H}_5\text{OH}$  instead of  $\text{MgCl}_2 \cdot 2.1 \text{ C}_2\text{H}_5\text{OH}$ .

Such catalyst component was used with dicyclopentyldimethoxysilane as external donor and with triethylaluminium (TEAL). The weight ratio TEAL/catalyst component was 5; the weight ratio TEAL/external donor was 4. Other operative conditions and the characteristics of the produced polymers are indicated in Table.

The catalyst was sent to the gas phase polymerisation apparatus. The latter comprised two interconnected cylindrical reactors, riser 1 and downcomer 2. Fast fluidisation conditions were established in reactor 1 by recycling gas from the gas-solid separator.

The way to differentiate the gas composition in the two reactor legs was the "barrier" feed. This stream was propylene fed in the larger upper part of the downcomer.

The polymer thus obtained is subjected to extrusion/granulation twin screw extruder ( $L/D=35$ ) in the presence of about 0.05% by weight of Luperox<sup>TM</sup> 101 (2,5-dimethyl-2,5-di-*t*-butylperoxy)esano and 0.05% by weight of calcium stearate and 0.15% by weight of Irganox<sup>®</sup> B215 (50 wt% of pentaerythryl-tetrakis[3(3,5-di-*tert*-butyl-4-hydroxyphenyl)]propionate and 50 wt% of bis(2,4-di-*tert*-butylphenyl)phosphite) marketed by Ciba-Geigy. The operative conditions are as follows: head temperature 250°C, melt temperature 239°C and output 21 kg/hour.

The polymerisation conditions and the features of the polymer before and after chemical degradation are reported in Table 1.

#### Comparative examples 1c and 2c

Conventional propylene homopolymers for spunbonding processing having the features reported in Table 1 are subjected to chemical degradation at the same conditions and with

the same agent as examples 1 and 2. The main properties of the degraded homopolymers are reported in Table 1.

Table 1

Example	1c	2c	1	2
1 <sup>st</sup> component				
Temperature °C	-	-	85	85
Split wt%	-	-	50	50
$C_2^-/(C_2^-+C_3^-)$ mol/mol	-	-	0.011	0.011
2 <sup>nd</sup> component				
Split wt%	-	-	50	50
$C_2^-/(C_2^-+C_3^-)$ mol/mol	-	-	0.1	0.1
Properties of precursor polymer composition				
MFR "L" g/10'	1.7	1.7	2.3	2.4
Polydispersity Index	3.6	3.6	3.7	3.7
Xylene-soluble fraction wt%	4.0	4.0	2.2	2
Xylene-insoluble fraction wt%	96	96	97.8	98
Ethylene content wt%	0	0	1.3	1.4
Melting temperature °C	163	163	158	158
Xylene-soluble fraction at the 25-95 °C temperature wt%	11.8	11.8	32.7	29.1
Properties of the visbroken polymer composition				
MFR "L" g/10'	25	28	25	27
P.I.	2.4	2.4	2.4	2.6
Xylene-soluble fraction at the 25-95 °C temperature wt%	13.9	-	33.3	33.3
TREF 25-95°C/xylene-soluble fraction ratio	3.5	-	15.1	16.6

Notes:

1<sup>st</sup> component = polymer produced in riser; 2<sup>nd</sup> component = polymer produced in downcomer;  
Split = amount of polymer produced;  $C_2^-$  = ethylene;  $C_3^-$  = propylene;  $C_2^-/(C_2^-+C_3^-)$  = monomer feed ratio.

Examples 3 and 4 and comparative example 3c

The polymers of examples 1 and 2 and comparative example 1c are spun in a Leonard 25 spinning pilot line with length/diameter ratio of the screw of 25, screw diameter of 25 mm and compression ratio of 1:3. The line is marketed by Costruzioni Meccaniche Leonard-Sumirago (VA)).

Table 2 reports the spinning characteristics and the properties of the filaments thus produced.

Table 2

Example		3c	3	4
Operative conditions				
Hole Diameter	mm	0,6	0.6	0.6
Output per hole	g/min	0.6	0.6	0.6
Hole number in the die		37	37	37
Die temperature	°C	250	250	250
Melt Temperature	°C	258	258	258
Take up speed	m/min	2700	2700	2700
Properties of the fibre				
Titre single fibre	dtex	2.25	2.3	2.35
Tenacity	cN/tex	21.1	26.8	24.6
Elongation at break	%	220	160	235

With respect to the filament of comparative example 3c, the filaments according to the present invention show increased values of the tenacity. The capability to produce filaments of higher tensile strength can be used advantageously for the production of spunbond non-woven fabrics.

#### Example 5 and comparative example 4c

The polymer of example 1 and comparative example 1c are processed in a spunbond line. The trials are conducted at full line capacity.

The non-woven fabrics thus produced have a weight of 15 g/m<sup>2</sup>.

#### Example 6

Example 5 is repeated excepted that the calender temperature is decreased by 5° C.

#### Example 7

Example 6 is repeated except that the filaments are processed under more severe conditions by increasing the pressure drag air.

In Table 3 the operative conditions and the properties of the non-woven fabrics obtained in examples 5 to 7 and comparative example 4c are reported.

Table 3

Example		4c	5	6	7
Operative conditions					
Temp. Melt	°C	225	225	225	225
Air jet pressure	mbar.	120	120	120	135
Output per hole	g/min	0.6	0.6	0.6	0.6
Temp. Smooth Calander	°C	168	168	163	163
Temp. Embossed Calander	°C	170	170	165	165
Properties of the filament and of the non-woven fabric					
Filament titre	dtex	2.9	3.0	2.9	2.8
Fabric weight	g/m <sup>2</sup>	15	15	15	15
Tenacity MD	N	26.1	33.2	31.8	34.3
Tenacity CD	N	7.6	8.9	7.9	8.2
Elongation at break MD	%	35	40	40	45
Elongation at break CD	%	55	65	60	65
Softness <sup>1)</sup>	1,2,3,4,5	3	4	5	5

<sup>1)</sup> Softness ranking: 1=bad; 2=lower than reference; 3= equal to reference; 4= better than reference; 5= Very good

The above data show that the filaments of example 3 have good processability, high consistency and no breakage under all tested conditions. The thus produced non-woven fabrics according to the present invention show higher softness and higher fabric tenacity than the comparative non-woven fabric.

#### Example 8 and comparative example 5c

The polymers of example 2 and comparative example 2c are tested in a spunbond line in the same conditions.

Table 4 summarises the main conditions of the extruder, spinning and calendering.

The non-woven fabric of example 8 shows higher tenacity in both machine and cross direction and lower elongation at break than the non-woven fabric of comparative example 5c

#### Example 9

The polymer of example 8 is spun at higher stretching air and lower calendering temperature (-5°C) than the polymer of example 8. No spinnability and process problems are experienced.

The non-woven fabric thus produced shows higher values of the tenacity in both MD and CD, still maintaining good values of the elongation at break in both MD and CD, than the

non-woven fabric of example 8. The polymer of comparative example 5c processed at the same calendering temperature leads to a non-woven fabric with poor tenacity.

Table 4

Example		5c	8	9
Temperature polymer melted in the filter	°C	240	240	240
Output per hole	g/min	0.39	0.39	0.39
Hole diameter	mm	0.45	0.45	0.45
Air jet pressure	MPa	15.19	15.19	16.17
Calendering temperature smooth roll	°C	139	139	134
Calendering temperature embossed roll.	°C	139	139	134
Properties of the filament and of the non-woven				
Filament titre	dtex	1.8	1.8	1.6
Non-woven weight	g/m <sup>2</sup>	16.3	16	16.5
Tenacity MD	N/5cm	33.2	33.5	35.4
Elongation at break MD	%	40	45	45
Tenacity CD	N/5cm	27.3	27.9	30.1
Elongation at break CD	%	35	45	45

Table 4 shows that the non-woven fabric according to the present invention has an increase of elongation at break and equivalent or even better tenacity in comparison with the comparative non-woven fabric and also has a good processability.

#### Example 10

The polymer is prepared and extruded as in example 1 except that in the polymerisation apparatus the two interconnected cylindrical reactors are downcomer 1 and riser 2 and no peroxide is added during the extrusion.

The polymerisation conditions and the main features of the polymer are reported in Table 5.

#### Comparative example 6c

Conventional propylene homopolymers for spunbonding processing having the features reported in Table 5 are subjected to chemical degradation just as in example 1, amounts and types of peroxide and additives being the same as in example 1. The main properties of the precursor and of the granulated polymers are reported in Table 5.

Table 5

Example	6c	10
1 <sup>st</sup> component		
Temperature °C	75	80
Split wt%	50	50
$C_2/(C_2+C_3)$ mol/mol	0	0.002
2 <sup>nd</sup> component		
Split wt%	50	50
$C_2/(C_2+C_3)$ mol/mol	0	0.02
Properties of precursor polymer composition		
MFR "L" g/10'	1.7	36.3
Polydispersity Index	3.6	3.35
Xylene-soluble fraction wt%	4.0	3.6
Xylene-insoluble fraction wt%	96.0	96.4
Ethylene content wt%	0	1.2
Melting temperature °C	163	156.8
Soluble fraction at the 25-95 °C temperature wt%	12.3	32.8
Properties of the polymer composition after granulation		
MFR "L" g/10'	32	36.4
P.I.	2.6	3.3
Ethylene content wt%	0	1.2
Melting temperature °C	163	156.8
Xylene-soluble fraction wt%	4.1	3.6
Xylene-soluble fraction at the 25-95 °C temperature wt%	13.9	32.8
TREF 25-95°C/xylene-soluble fraction ratio	3.5	9.1

Note: 1<sup>st</sup> component = polymer produced in downcomer; 2<sup>nd</sup> component = polymer produced in riser;

Split = amount of polymer produced;  $C_2$  = ethylene;  $C_3$  = propylene;  $C_2/(C_2+C_3)$  = monomer feed ratio.

Example 11 and comparative example 7c

The polymers of 10 and comparative example 6c are tested in a spunbond line in the same conditions.

The trials are conducted at standard line capacity.

The non-woven fabrics thus produced have a weight of 35 g/m<sup>2</sup>.

In Table 6 the operative conditions and the properties of the non-woven fabrics obtained in examples 11 and comparative example 7c are reported.

Table 6

Example		7c	11
Operative conditions			
Temp. Melt	°C	225	225
Air jet pressure	Mbar	800	800
Quenching air temperature	°C	15	15
Output per hole	g/min	0.5	0.5
Extruder pressure	Bar	80	80
Extruder screw revolution	Rpm	40	40
Belt speed	m/min	45	45
Temp. Smooth Calender	°C	139	139
Temp. Embossed Calender	°C	142	142
Properties of the filament and of the non-woven fabric			
Filament titre	dtex	2.0	2.0
Fabric weight	g/m <sup>2</sup>	35	35
MD Tenacity	N/5cm	85.1	93.7
CD Tenacity	N/5cm	56.3	64.0
MD Elongation at break	%	70.9	116.1
CD Elongation at break	%	68.8	117

The polymer of example 10 shows good processability, high consistency and no breakages has been experienced during the test. The thus produced non-woven fabrics according to the present invention show higher elongation with higher fabric tenacity than the comparative non-woven fabric.

25. Sep. 2002

## Claims

1. A fibre for spun bonding comprising a propylene polymer composition (A) having an MFR L (1) value from 6 to 60 g/10 min and being selected from:
  - i) a crystalline propylene random copolymer containing at least 0.8% by weight of ethylene and optionally one or more C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefins; the said polymer having a melting temperature of 155° C or higher, a content of fraction soluble in xylene at room temperature (about 25° C) lower than 4% by weight, preferably lower than 3% by weight, and a value of the ratio of the polymer fraction collected at the temperature range from 25° to 95° C by temperature rising elution fractionation with xylene to the xylene soluble fraction at room temperature, higher than 8; and
  - ii) a crystalline propylene polymer blend having a melting temperature of 153° C or higher, a content of fraction soluble in xylene at room temperature lower than 10% by weight; the said blend containing at least 0.64 wt% of ethylene recurring unit and comprising (percent by weight):
    - I) 20-80%, preferably from 30-70%, of a crystalline propylene homopolymer or crystalline propylene random copolymer containing up to 1.5% by weight of ethylene; and
    - II) 20-80%, preferably from 30-70%, of a crystalline propylene random copolymer selected from:
      - IIa) a copolymer of propylene with 0.8 to 9% by weight of ethylene; provided that the difference in the ethylene content between polymer (I) and polymer (IIa) be at least 0.8 percentage unit with respect to the weight of the (co)polymer concerned;
      - IIb) a copolymer of propylene with 1.5 to 18% by weight of a C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefin and optionally ethylene; provided that the difference in the comonomer content between polymer (I) and polymer (IIb) be at least 1.5 percentage units with respect to the weight of the (co)polymer concerned; and
      - IIc) a mixture of copolymer (IIa) and copolymer (IIb).
2. The fibre of claim 1 wherein composition (A) is polymer blend (ii) having a melting temperature of 155° C or higher, a content of fraction soluble in xylene at room



temperature lower than 5% by weight and a value of the ratio of the polymer fraction collected at the temperature range from 25° to 95° C by temperature rising elution fractionation with xylene to the xylene soluble fraction higher than 8; said blend (ii) comprising (percent by weight):

I) 20-80%, preferably 30-70%, of a crystalline propylene homopolymer and/or crystalline propylene random copolymer containing up to 1.5% by weight of ethylene; and

II) 20-80%, preferably 30-70%, of a crystalline random copolymer selected from:

IIa) a copolymer of propylene with 0.8 to 5% by weight of ethylene; provided that the difference in the ethylene content between polymer (I) and polymer (IIa) be at least 0.8 percentage unit with respect to the weight of the (co)polymer concerned;

IIb) a copolymer of propylene with 1.5 to 12% by weight of a C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefin and optionally ethylene; provided that the difference in the comonomer content between polymer (I) and polymer (IIb) be at least 1.5 percentage units with respect to the weight of the (co)polymer concerned; and

IIc) a mixture of copolymer (IIa) and copolymer (IIb).

3. The fibre of claims 1 and 2 wherein composition (A) is obtainable by subjecting to chemical degradation a precursor polymer composition (B) having an MFR L value (MFR L (2)) of from 0.5 to 6 g/10 min, preferably from 0.5 to 3.5 g/10 min, provided that the ratio MFR L (1) to MFR L (2) be from 1.5 to 60, preferably from 6 to 30.
4. The fibre of claims 1 to 3 wherein the MFR L (1) values range from 10 to 40 g/10 min.
5. The fibre of claims 1 to 4 wherein the difference in the ethylene content between polymer (I) and polymer (IIa) is at least 1 percentage unit with respect to the weight of the (co)polymer concerned.
6. A melt spin process for the production of the fibres according to claims 1 to 5 characterised in that it is subjected to the process a propylene polymer composition (A) having MFR L (1) values from 6 to 60 g/10 min and being selected from:
  - i) a crystalline propylene random copolymer containing at least 0.8% by weight of ethylene and optionally one or more C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefins; the said polymer having a melting temperature of 155° C or higher, a content of fraction soluble in xylene at room temperature lower than 4% by weight, preferably lower than 3% by

weight, and a value of the ratio of the polymer fraction collected at the temperature range from 25° to 95°C by temperature rising elution fractionation with xylene to the xylene soluble fraction at room temperature, higher than 8; and

- ii) a crystalline propylene polymer blend having a melting temperature of 153° C or higher, a content of fraction soluble in xylene at room temperature lower than 10% by weight; the said blend containing at least 0.64 wt% of ethylene recurring unit and comprising (percent by weight):

I) 20-80%, preferably from 30-70%, of a crystalline propylene homopolymer or crystalline propylene random copolymer containing up to 1.5% by weight of ethylene; and

II) 20-80%, preferably from 30-70%, of a crystalline propylene random copolymer selected from:

IIa) a copolymer of propylene with 0.8 to 9% by weight of ethylene; provided that the difference in the ethylene content between polymer (I) and polymer (IIa) be at least 0.8 percentage unit with respect to the weight of the (co)polymer concerned;

IIb) a copolymer of propylene with 1.5 to 18% by weight of a C<sub>4</sub>-C<sub>10</sub> α-olefin and optionally ethylene; provided that the difference in the comonomer content between polymer (I) and polymer (IIb) be at least 1.5 percentage units with respect to the weight of the (co)polymer concerned; and

IIc) a mixture of copolymer (IIa) and copolymer (IIb);

7. A propylene polymer blend having an MFR L (1) value from 6 to 60 g/10 min, the said composition comprising (percent by weight):

I) 20-80%, preferably from 30-70%, of a crystalline propylene homopolymer or crystalline propylene random copolymer containing up to 1.5% by weight of ethylene; a melting temperature of 155° C or higher, a content of fraction soluble in xylene at room temperature (about 25° C) lower than 4% by weight, preferably lower than 3% by weight, and a value of the ratio of the polymer fraction collected at the temperature range from 25° to 95°C by temperature

rising elution fractionation with xylene to the xylene soluble fraction at room temperature, higher than 8; and

II) 20-80%, preferably from 30-70%, of a crystalline propylene random copolymer selected from:

IIa) a copolymer of propylene with 0.8 to 9% by weight of ethylene; provided that the difference in the ethylene content between polymer (I) and polymer (IIa) be at least 0.8 percentage unit with respect to the weight of the (co)polymer concerned;

IIb) a copolymer of propylene with 1.5 to 18% by weight of a C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefin and optionally ethylene; provided that the difference in the comonomer content between polymer (I) and polymer (IIb) be at least 1.5 percentage units with respect to the weight of the (co)polymer concerned; and

IIc) a mixture of copolymer (IIa) and copolymer (IIb);

said polymer composition being obtainable by way of chemical degradation of a precursor polymer composition (A) having an MFR L value (MFR L (2)) of from 0.5 to 6 g/10 min, provided that the ratio MFR L (1) to MFR L (2) be from 1.5 to 60.

8. A process for the preparation of the polymer blend of claim 7 comprising the following stages:

- 1) preparing the previously said precursor composition (B) by polymerising the monomers in one or more sequential steps, operating in each step in the presence of the polymer formed and the catalyst used in the preceding step, and dosing the molecular weight regulator (preferably hydrogen) in such amounts as to obtain an MFR L (2) value for the precursor composition of from 0.5 to 6 g/10 min, preferably from 0.5 to 3.5 g/10 min; and
- 2) subjecting the precursor composition (B) obtained in 1) to a degradation treatment with a degradation ratio, in terms of ratio MFR L (1) to MFR L (2), of from 1.5 to 60.

9. A spunbonded non-woven fabric comprising the fibres of claim 1.

10. A multilayered non-woven fabric wherein at least one layer is formed from the fibres of claim 1.

25. Sep. 2002

### Abstract

A fibre for spun bonding comprising a propylene polymer composition (A) having an MFR L (1) value from 6 to 60 g/10 min and being selected from (i) a crystalline propylene random copolymer containing at least 0.8% by weight of ethylene; the said polymer having a melting temperature of 155° C or higher, a content of fraction soluble in xylene at room temperature lower than 4% by weight, and a value of the ratio of the polymer fraction collected at the temperature range from 25° to 95°C by temperature rising elution fractionation with xylene to the xylene soluble fraction at room temperature, higher than 8; and (ii) a crystalline propylene polymer blend having a melting temperature of 153° C or higher, a content of fraction soluble in xylene at room temperature lower than 10% by weight; the said blend containing at least 0.64 wt% of ethylene recurring unit and comprising (percent by weight) (I) 20-80 wt% of a crystalline propylene homopolymer or crystalline propylene random copolymer containing up to 1.5% by weight of ethylene and II) 20-80 wt% of a crystalline random copolymer of propylene with ethylene or a C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefin.

A polymer blend having an MFR L (1) value from 6 to 60 g/10 min, the said composition comprising (I) 20-80 wt% of a crystalline propylene homopolymer or crystalline propylene random copolymer containing up to 1.5% by weight of ethylene; a melting temperature of 155° C or higher, a content of fraction soluble in xylene at room temperature lower than 4% by weight, and a value of the ratio of the polymer fraction collected at the temperature range from 25° to 95°C by temperature rising elution fractionation with xylene to the xylene soluble fraction at room temperature, higher than 8; and (II) 20-80 wt% of a crystalline random copolymer propylene with ethylene or a C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefin. The said polymer composition is obtainable by way of chemical degradation of a precursor polymer composition (A) having an MFR L value (MFR L (2)) of from 0.5 to 6 g/10 min, provided that the ratio MFR L (1) to MFR L (2) is from 1.5 to 60.

Non-woven fabrics that are prepared with the said fibres are useful for coverstock and diapers.

**This Page is Inserted by IFW Indexing and Scanning  
Operations and is not part of the Official Record**

**BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ **BLACK BORDERS**
- ☐ **IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- ☐ **FADED TEXT OR DRAWING**
- ☐ **BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- ☐ **SKEWED/SLANTED IMAGES**
- ☐ **COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- ☐ **GRAY SCALE DOCUMENTS**
- ☒ **LINES OR MARKS ON ORIGINAL DOCUMENT**
- ☐ **REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- ☐ **OTHER:** \_\_\_\_\_

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.**